ENGINEERING EXPERIMENT STATION
of the Georgia Institute of Technology
Atlanta, Georgia

FINAL REPORT

PROJECT NO. A-136

EFFECT OF VARIATION OF TEMPERATURE AND PRESSURE ON COMPOSITION OF ALLOYS

by

WILLIAM M. SPICER

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Contract AF 18(600)-974

AIR RESEARCH AND DEVELOPMENT COMMAND
Project No. R-355-20-14

January 1, 1954 through December 31, 1955

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I ABSTRACT

Experiments were performed on several solid alloys in an attempt to show that concentration gradients might develop as a result of a pressure gradient or a temperature gradient.

The results on the pressure effect were inconclusive. However, the results on the temperature effect, especially those on the Pb-14.8 per cent Sn and the Al-21.9 per cent Zn alloys, indicate strongly that a concentration gradient did develop as a result of the temperature gradient. In the case of the Pb-14.8 per cent Sn alloy, the percentage tin reached a maximum of 15.71 per cent near the hot end and a minimum of 13.84 per cent near the cold end. With the Al-21.9 per cent Zn alloy, the percentage of zinc attained a maximum of 25.57 per cent and a minimum of 18.12 per cent in one of the three runs.

II PURPOSE

The purpose of Project No. A-136 under Contract No. AF 18(600)-974 entitled "Study of the Effect of the Variation of Temperature and Pressure on the Composition of Alloys", is to determine experimentally the answers to the following questions:

(1) If a portion of an alloy of uniform composition is maintained at a temperature different from that of the remainder of the alloy, will the composition gradually change as a result of this temperature difference?

(2) Similarly, if a portion of such an alloy, at uniform temperature, is subjected to a high pressure, will the composition gradually change as a result of this pressure difference?

III THEORETICAL FOUNDATIONS

A. The Pressure Effect

Consider a piece of a homogeneous alloy at some temperature and pressure as divided into two portions. A condition of equilibrium is that the fugacity of the i^{th} component is the same in the two portions, i.e.,

$$f_i = f_i' \tag{1}$$

where one portion is referred to as the prime portion. We could write from Equation 1,

$$\ln f_i = \ln f_i' \tag{2}$$

Now, if conditions are changed, but the equilibrium maintained,

$$d\ln f_i = d\ln f_i'. \tag{3}$$

Suppose the change in conditions consists of increasing the pressure on the primed portion, while maintaining constant temperature. As the pressure changes, the concentrations of the components of the alloy will change and Equation 3 becomes

$$\left(\frac{\partial \ln f_i}{\partial N_i}\right)_{P,T} dN_i = \left(\frac{\partial \ln f_i'}{\partial N_i'}\right)_{P,T} dN_i' + \left(\frac{\partial \ln f_i'}{\partial P}\right)_{T,N_i'} dP \tag{4}$$

where N is the mole fraction.

Equation 4 applies, of course, to each component of the alloy. For a binary alloy, for example,

$$\left(\frac{\partial \ln f_i}{\partial N_i}\right)_{P,T} dN_1 = \left(\frac{\partial \ln f_1'}{\partial N_1'}\right)_{P,T} dN_1' + \left(\frac{\partial \ln f_1'}{\partial P}\right)_{T,N_1'} dP \qquad (5)$$

$$\left(\frac{\partial \ln f_2}{\partial N_2}\right)_{P,T} dN_2 = \left(\frac{\partial \ln f_2'}{\partial N_2'}\right)_{P,T} dN_2' + \left(\frac{\partial \ln f_2'}{\partial P}\right)_{T,N_2'} dP \qquad (6)$$

In general, $\left(\frac{\partial \ln f_i}{\partial P}\right)_{T,N_i} = \frac{\bar{V}_i}{RT}$, where $\bar{V}_i$ is the partial molal volume of the i^{th} component in the solution. However, the derivative $\left(\frac{\partial \ln f}{\partial N}\right)_{P,T}$ cannot be further reduced without additional knowledge about the particular solid solution under consideration. If we assume, as a first approximation, that f is proportional to N, then

$$\left(\frac{\partial \ln f}{\partial N}\right)_{P,T} = \frac{1}{N}$$

and

$$\left(\frac{\partial \ln f}{\partial N}\right)_{P,T} dN = \frac{dN}{N} = d\ln N.$$

Equations 5 and 6 now reduce to

$$d\ln N_1 = d\ln N_1' + \frac{\bar{V}_1}{RT} dP \qquad (7)$$

or

$$\left(\frac{\partial \ln \frac{N_1}{N_1'}}{\partial P}\right)_T = \frac{\bar{V}_1}{RT} \tag{7}$$

and

$$d\ln N_2 = d\ln N_2' + \frac{\bar{V}_2}{RT}\, dP$$

or

$$\left(\frac{\partial \ln \frac{N_2}{N_2'}}{\partial P}\right)_T = \frac{\bar{V}_2}{RT} \,. \tag{8}$$

Subtracting Equation 8 from Equation 7, we get

$$\left(\frac{\partial \ln \frac{N_1}{N_1'}\frac{N_2'}{N_2}}{\partial P}\right)_T = \frac{\bar{V}_1 - \bar{V}_2}{RT} \,. \tag{9}$$

Little is known about the partial molal volumes of metals in solid solutions. However, again as a first approximation, we might assume that the difference in the partial molal volumes is equal to the difference in the molal volumes of the pure components. For example, tin with an atomic weight of 118.7 has a density of 7.3 and an atomic volume of approximately 16 cc. The corresponding figures for copper are 63.6, 8.9, and 7 cc. Thus, at say 1,000° K, the right-hand side of Equation 9 is approximately

$$\frac{16-7}{82.05 \times 1000} \approx -1 \times 10^{-4} \text{ atm.}^{-1},$$

and Equation 9 becomes

$$\left(\frac{\partial \ln \frac{N_{Cu}}{N'_{Cu}} \frac{N'_{Sn}}{N_{Sn}}}{\partial P}\right)_T = -1 \times 10^{-4} \text{ atm.}^{-1}. \qquad (10)$$

If we assume that this derivative is constant, we can integrate and obtain

$$\ln \frac{N_{Cu}}{N_{Sn}} \frac{N'_{Sn}}{N'_{Cu_2}} - \ln \frac{N_{Cu}}{N_{Sn}} \frac{N'_{Sn}}{N'_{Cu_1}} = -1 \times 10^{-4} \times \Delta P. \qquad (11)$$

Suppose that in the original alloy $N_{Cu} = N_{Sn} = 0.5$, then

$$\ln \frac{N_{Cu}}{N_{Sn}} \frac{N'_{Sn}}{N'_{Cu_1}} = \ln 1 = 0.$$

If, for example, ΔP is taken as 10,000 atm., Equation 11 reduces to

$$\ln \frac{N_{Cu}}{N_{Sn}} \frac{N'_{Sn}}{N'_{Cu_2}} = -1. \qquad (12)$$

Suppose now that the portion of the alloy which was not compressed is so large relative to the compressed portion that its composition does not change, i.e.,

$$\frac{N_{Cu}}{N_{Sn}} = 1, \text{ then } \ln \frac{N'_{Sn}}{N'_{Cu}} = -1$$

and

$$\log \frac{N'_{Sn}}{N'_{Cu}} = -0.429 = \bar{1}.572,$$

therefore,

$$\frac{N'_{Sn}}{N'_{Cu}} = 0.37.$$

Thus the tin, since it occupies the larger volume, will escape from the compressed portion and the $\frac{N'_{Sn}}{N'_{Cu}}$ will decrease from its original value of unity to an equilibrium value of only 0.37.

1. The Effect of Pressure on Rate of Diffusion

High pressures might be expected to decrease diffusion rates; since, when a material is compressed, the atoms are forced closer together, and their freedom of motion decreased. Little experimental work has been done in this field (2). According to Cohen and Bruins (3), an increase of 1,500 atmospheres decreases the diffusion rate of cadmium in mercury by 5 per cent at 20° C. Radavich and Smoluchowski (4) reported that for the aluminum-copper system at 500° C, a pressure of 7,000 $\frac{kg}{cm^2}$ decreases the diffusion by 30 per cent. However, Johnson and Adams (5) found some evidence that pressure increases the rate of diffusion.

B. The Temperature Effect

If a portion of an alloy is held at a temperature different from that of another portion, a concentration difference may develop corresponding to the temperature difference. There are two possible and quite different reasons for this:

(1) The so-called Ludwig-Soret Effect (1).

(2) An effect due to a change in solubility with temperature. To illustrate this, consider the silver-copper diagram shown in Figure 1. Suppose a portion of an alloy containing somewhat less than 8.8 weight per cent copper is maintained at 800° C while another portion is maintained at say 300° C. The 800° C portion consists of an unsaturated solution of copper in silver, while the 300° C portion consists of a saturated solution of copper in silver along with either an excess silver or a saturated solution of silver in copper (which of these is not clear from the diagram). In either case it would appear that copper should move from the cool to the hot portion.

A preliminary study of a number of systems was made in order to determine the most favorable one on which to concentrate the main effort. Alloys investigated by us were 98.9 per cent Mg-1.1 per cent Mn, 79 per cent Al-21 per cent Zn, 86 per cent Pb-14 per cent Sn, 90 per cent Cu-10 per cent Sn, 92.5 per cent Cu-7.5 per cent Ag (sterling silver), 99.5 per cent Cu-0.5 per cent Cr, and 99.6 per cent Cu-0.4 per cent Ni.

Also after the work of Darken and Oriani (6) appeared, a run was made on 95 per cent Au-5 per cent Cu alloy in an attempt to reproduce their results. Since Darken and Oriani successfully demonstrated the existence of the Ludwig-Soret effect, it was decided to devote our main effort to an investigation of whether a concentration difference would develop in different portions of an alloy as a result of a difference in solubility at different temperatures.

From a study of the phase diagrams of the various alloys, it appeared that the Al-Zn system would be most favorable for this investigation since it shows a very rapid change in solubility with temperature (see Figure 2). Using

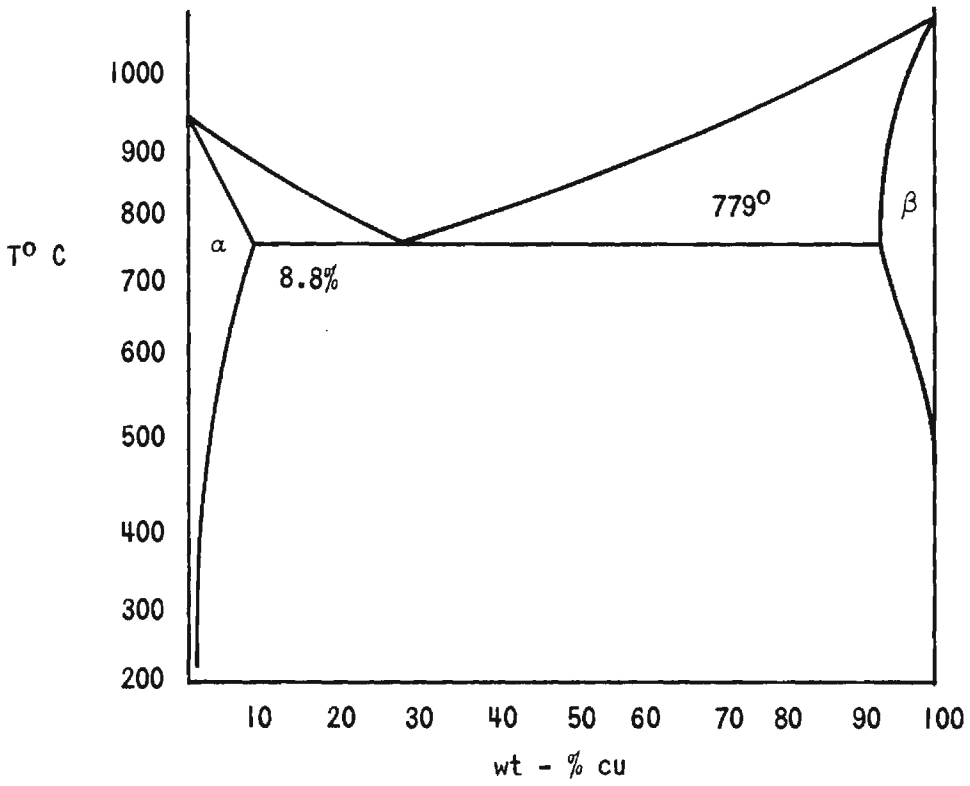

Figure 1. Silver-Copper Phase Diagram.

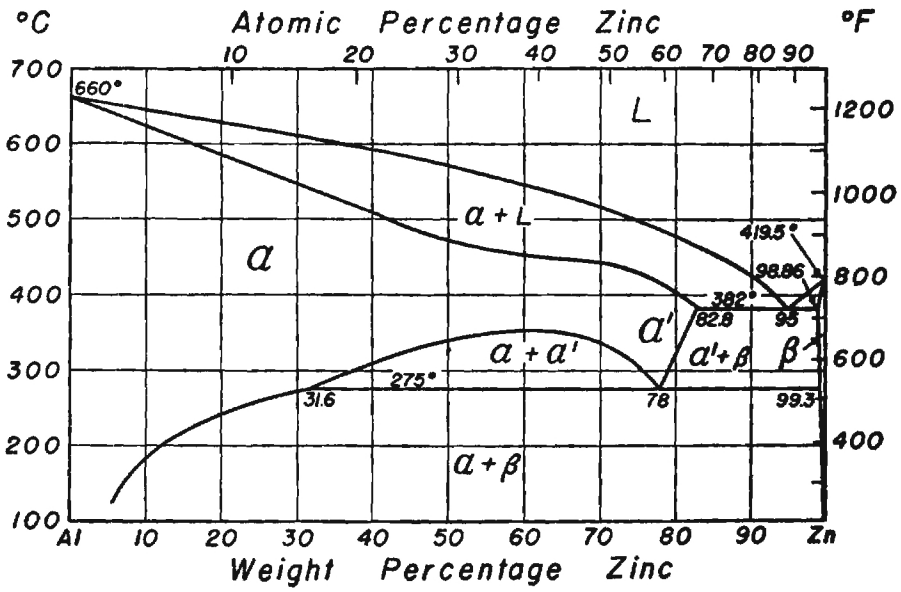

Figure 2. Aluminum-Zinc Phase Diagram.

a 21 per cent zinc alloy, for example, it will be noted that at 100° C the equilibrium alloy consists of two phases, one being essentially pure zinc and the other being a solid solution containing only about 5 per cent zinc. However, if the temperature is raised to 382° C, the solubility of zinc in aluminum rises from 5 per cent to 82.8 per cent. From the diagram it appears that the boundary separating the unsaturated from the saturated solution for the 21 per cent zinc alloy is at approximately 250° C. Thus it would appear that if one end of a rod of this alloy were maintained at some temperature below 250° C, and the other end at some temperature above 250° C, zinc should migrate from the low temperature saturated solution to the high temperature unsaturated solution. In short, a concentration difference should develop as a result of the temperature difference.

Preliminary results confirmed that this is a favorable system for this study.

(If everything pointed to this system as the best, why were others investigated? The answer is that it was difficult to obtain this alloy since it is not commercially available. It was finally made especially for us and contributed by the Aluminum Company of America.)

IV PRIOR WORK

So far as we have been able to determine no work has been done on either the pressure effect or on the change of composition due to the effect of temperature on solubility. The Ludwig-Soret effect has, however, been investigated to some extent. The earliest work was done by Ballay (1) using alloys of lead-thallium, lead-tin, platinum-rhodium, and copper-aluminum. He obtained inconclusive results. The most recent work is that of Darken and Oriani (6). They demonstrated the effect with α-Fe-N, α-Fe-C, and Au-Cu. They also review the literature.

Winter and Drickamer have just published some results on liquid alloys. (December, 1955) (11).

V EXPERIMENTAL

A. Apparatus

1. The Pressure Effect

The following equipment was constructed to exert the necessary high pressure on the sample under study. A steel cylinder 2 inches long and 1 inch in diameter was made. Both sides were flattened at the bottom in order to hold it in a vice. A slot 1/2-inch wide and 1/4-inch high was cut through the cylinder halfway up its side. In the center of the upper half of the cylinder, a 9/16-inch diameter screw was fitted. The sample was placed in the slot under the screw. By tightening the screw, pressure was exerted on that part of the sample under it. It was estimated that the force on the sample under the screw was 15T, where T was the torque in inch-pounds. Thus, if a pull of 100 pounds were used with a 10-inch wrench, the force would be 15,000 pounds. The bottom of the screw was only 7/32-inch in diameter. Its area, from which the threads had been cut, was, therefore, approximately 0.04 sq. in. and the pressure on the sample was $\frac{15,000}{0.04}$ = 375,000 psi = $\frac{375,000}{15}$ = 25,000 atm. (Since we are interested at first in merely detecting the pressure effect, we are not attempting an accurate calculation of the pressure.)

In order to maintain the sample under pressure at the chosen high constant temperature, a 550-watt electric tube furnace was used. The temperature of the furnace was controlled by means of a Variac connected to a constant-voltage transformer. The cylinder holding the sample was placed in a 1 1/4-inch Pyrex tube which fitted snugly into the furnace. One end of the Pyrex tube, which protruded about a foot out of the furnace, was closed with a ground-glass joint and fitted with a three-way stopcock. Through this stopcock the furnace could be evacuated and argon introduced.

Before introducing the sample into the furnace, tests were made to determine both the constant temperature zone of the furnace and the Variac settings which produced certain chosen temperatures. In order to do this, we used a probe consisting of a steel cylinder, approximately the same size as the pressure cylinder, in which was placed the junction of a chromel-alumel thermocouple.

The main criticism of this method for subjecting the alloys to pressure (i. e., by means of a bolt) is when pressure is relieved some of the alloy creeps from under the bolt. In view of this, we have always tightened the bolt several times during a given run. However, in order to avoid this difficulty another apparatus has been constructed which will exert a continuous constant force on the alloy even if the alloy does creep. The force is exerted by a large steel ingot weighing 1,255 pounds. By using a small rod between the ingot and the alloy, a high pressure can be exerted on a portion of the alloy. (See Figure 3.)

2. The Temperature Effect

In order to study the temperature effect, two different type setups were used. In the first of these, each end of the alloy rod is threaded and screwed into a somewhat larger steel cylinder, whose purpose is to add "body" in order to make it easier to maintain constant temperature. One cylinder and, therefore, one end of the alloy rod, is maintained at a relatively high temperature in an electric furnace, while the other end is cooled by means of a water condenser. This condenser is made of brass and is soldered to the end of the steel cylinder. The temperature of the hot end within the cylinder is measured by means of a thermocouple, that of the "cold" end is estimated to be

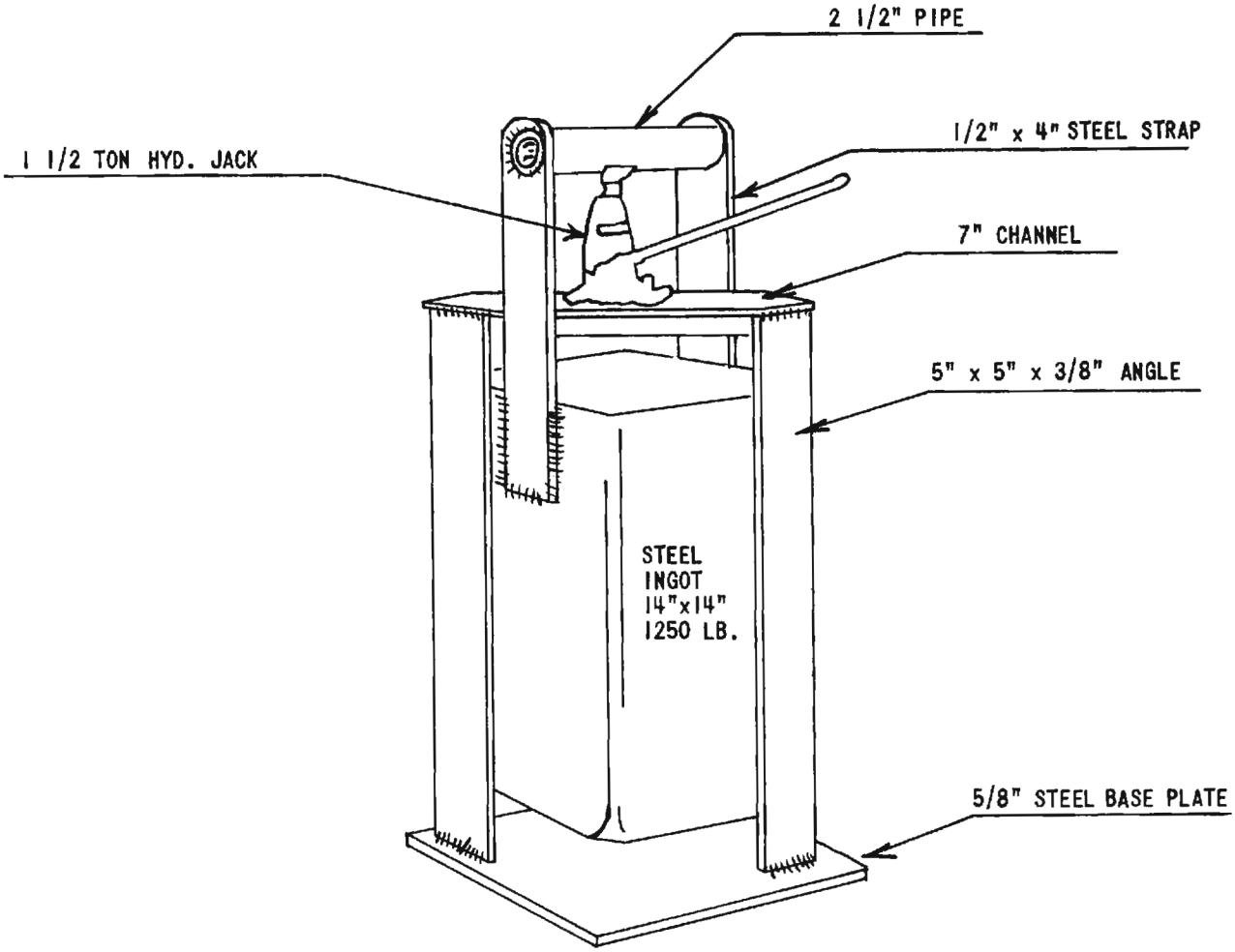

Figure 3. Pressure Exerting Apparatus.

slightly above that of boiling water. The setups are enclosed in evacuated glass, or, in case of the higher temperature ones, quartz tubes. This is the type setup that was used with the Al-Zn alloy.

The other type of apparatus used for the study of the temperature effect consists of a steel bell jar 8 inches in diameter and 15 inches high, resting on a brass base 12 inches square and 1/2-inch thick. A brass ring is soldered to the bottom of the bell jar in order to fit it more snugly to the base. The base stands on four steel legs 4 1/2-inches high. This space was left under the base in order to facilitate bringing out leads. In order to cool the base, a spiral of copper tube, through which water could be circulated, was soldered to it. Thermocouple wires as well as leads to heating coils also pass through the base. A rubber gasket is used between the base and the bell jar in order to obtain a vacuum seal. In order to subject a portion of a sample to a high temperature and another portion to a low temperature in this bell jar, the following arrangement was made. An aluminum block about 1/2-inch thick was polished quite smooth so that it made good contact with the cold brass base. In the center of this block a 1/4-inch diameter hole was drilled vertically almost to the bottom of the block. In this hole the 1/4-inch diameter rod sample which was to be subjected to test was fitted. This arrangement not only held the sample upright, but it also served to keep the lower end cool. Above the upper half of the alloy rod, a small resistance furnace made by releasing a coil of nichrome wire in 11-mm bore of a Sillimanite block 2 inches high and 1 1/4-inches square was held. The power furnished this coil was controlled by a Variac operated from a constant-voltage transformer. To accommo-

date the thermocouple junction for temperature measure, small holes were drilled in the alloy rod, one at the top and another near the bottom.

To evacuate the bell jar, a Cenco Megavac pump was used.

B. Results

1. The Pressure Effect

Using the apparatus in which the pressure is exerted on the sample by means of a screw, the following alloys, which are all homogeneous, solid solutions, were studied: Cu-10 per cent Sn, Cu-5 per cent Sn, Cu-2 per cent Sn, Cu-15 per cent In, Pb-15 per cent Sn, Al-10 per cent Mg, Al-24 per cent Zn.

The source of these alloys was as follows: the Cu-Sn alloys were made by the Southern Research Institute; the Cu-In alloy was made in our laboratory; the Al-Mg alloy was contributed by the Dow Chemical Company; the Al-Zn alloy was contributed by the Aluminum Company of America; and, the Pb-Sn alloy was obtained commercially.

(These alloys were chosen because of the considerable difference in the atomic diameters of their components. The atomic diameters are Cu-2.56 A, In-3.24 A, Sn-2.8 A, Pb-3.50 A, Al-2.86 A, Mg-3.20 A, An-2.66 A.)

Approximately every ten days during the runs the furnace was turned off, and, as soon as the samples had cooled, the screws were retightened in order to continue to maintain the pressure on the sample. Each time this was done, it was found that the same force that was used on the screw at the beginning of the run moved the screw from approximately 1/8 to 1/4 of the turn. It was realized, of course, that this continuous decrease in pressure was due mainly

to the plastic flow of the alloy from the high pressure region. This plastic flow will no doubt tend to obscure the effect which is sought.

In general, these experiments failed. The most encouraging results were obtained with the Cu-5 per cent Sn alloy which had been subjected to pressure for two months at 590° C as follows: (The copper was determined by electrolysis and the percentage of tin was obtained by difference.)

Location of the Sample Analyzed	Per Cent Cu	Per Cent Sn
Completely inside pressure zone	95.33	4.67
Just inside pressure zone	95.61	4.39
Just outside pressure zone	94.62	5.38
Completely outside pressure zone	94.60	5.40
Far outside pressure zone	94.41	5.59

These results appear quite convincing; they seem to indicate that the tin, the element with the larger atomic volume migrated, as theory predicted, from the region of high pressure. However, it is possible that these results were merely fortuitous since those obtained with the other systems, even the Cu-Sn ones, gave no such convincing picture. For example, the results on the other two Cu-Sn alloys, which had had essentially the same treatment as the above one, were as follows:

	2 Per Cent Sn Alloy		10 Per Cent Sn Alloy	
Location of the Sample Analyzed	% Cu	% Sn	% Cu	% Sn
Completely inside pressure zone	97.46	2.54	85.71	14.29
Just inside pressure zone	98.09	1.91	90.16	9.84
Just outside pressure zone	-----	----	85.58	14.42
Completely outside pressure zone	97.43	2.57	85.58	14.42
Far outside pressure zone	97.74	2.26	86.49	13.51

Using the dead-weight method of exerting pressure, the following systems were studied: phosphor-copper alloy containing 0.014 per cent P, Cu-1 per cent Pb (an eutectic mixture rather than a solid solution), and Cu-2 per cent Sn.

a. Phosphor-Copper Alloy. The details of the method will be illustrated by describing the run on the Cu-P system. The specimen consisted of the two halves of a disc 5/8-inch in diameter and approximately 1/8-inch thick. The pressure was exerted on 1/8-inch portions along the straight edge of the two halves by the 1,255 pound weight. (The area of the specimen supporting the weight was approximately 5/8 x 1/8 x 2 = 10/64 sq. inches. The pressure was, therefore, 1255 x 64/10 ~ 8000 pounds per sq. inch ~ 550 atm. When this pressure, instead of 10,000 atm, is sutstituted into Equation 11 (p.6), one finds that the $\frac{N'_{Sn}}{N'_{Cu}}$ should decrease from its original value of 1 to an equilibrium value of 0.945.) The temperature used was 300° C, and the time was 59 days. The sample was analyzed spectrographically using the 2553.25 A phosphorus line and the 2614.3 A copper line. No significant difference in the relative intensities of these lines was observed when that portion of the alloy that had been subjected to pressure was compared with a portion of the original alloy.

b. Copper-Lead Alloy. Copper and lead form an eutectic mixture (melting point, 326° C) and there is a large difference in their atomic volumes, that of lead being 18 cc per gram-atom and that of copper being only 7 cc per gram-atom. The large difference in atomic volume is important since the magnitude of the effect should be proportional to this difference and the eutectic type important since it would be expected that migration would take place more

rapidly in this type than in solid solution, since it could take place along grain boundaries.

A run was made on this system for 64 days at 240° C. The sample was then analyzed spectrographically using the 2663.2 A lead line and the 2997.4 A copper line with spark excitation. A portion of the original alloy was analyzed similarly at the same time. The results are given in Table I below. The first two columns are the transmission readings on the lead and copper lines, respectively, and the third column is the ratio of lead line intensity to that of the copper line.

TABLE I

TRANSMISSIONS Pb-2663.2 A	Cu-2997.4 A	RATIO $\frac{\text{Pb-2663.2}}{\text{Cu-2997.4}}$
	On Sample (Subjected to Pressure)	
66.0	17.9	0.38
47.0	12.7	0.45
39.4	10.4	0.46
57.2	20.7	0.48
	On Blank (Unpressed Specimen)	
35.7	45.2	1.2
50.5	46.5	0.93
50.5	37.7	0.79
52.4	38.0	0.76

It will be noted that although the results on the blank show considerable variation, they indicate that the percentage of lead in the blank is much higher

than in that portion of the sample which had been subjected to pressure. These results were encouraging.

Proof that lead had actually moved from the high-pressure zone to the low-pressure zone of the sample would require a similar analysis on the portion of the sample that was not subjected to the high pressure to show that lead had increased in concentration in this region. The small size of the specimen and its semi-circular shape prevented such an analysis. A new run, therefore, was made on a rectangular, rather than a semi-circular, specimen. This run was continued for 38 days. The results are given in Table II below.

TABLE II

TRANSMISSIONS		RATIO
Pb-2663.2	Cu-2997.4	$\frac{\text{Pb-2663.2}}{\text{Cu-2997.4}}$
	On Sample (Subjected to Pressure)	
51.0	23.9	0.58
51.0	19.5	0.52
67.8	29.1	0.48
61.7	28.0	0.53
	On Blank	
48.5	34.2	0.76
40.6	30.0	0.80
50.5	37.4	0.78
38.0	31.4	0.88
	On Sample (Not Subjected to Pressure)	
62.5	23.5	0.47
60.0	25.4	0.51
70.0	32.1	0.50
77.0	36.6	0.48

The results on the blank and on that portion of the specimen which had been subjected to pressure agreed fairly well with those on the first specimen, however, those on the uncompressed portion of the specimen were lower, rather than higher, than those of the blank. In fact, they were as low as those on the compressed portion. Thus, we conclude that although lead left the region of compression, it did not enter the region not under compression. On the contrary, lead also left this uncompressed region. This means that lead either evaporated or was removed by the molybdenum sheets that were in contact with the specimens while they were being subjected to pressure. That this latter was the case was proven by qualitative spectrographic analysis of one of these sheets which showed appreciable quantities of lead in the sheet. As mentioned previously, the Pb-Cu alloy is of the eutectic type, i. e., the lead is present as a separate phase. If it had been in solid solution, it likely would not have been removed by the molybdenum.

c. Copper-Tin Alloy. A run was made on this alloy for 197 days at 300° C. The sample was then analyzed spectrographically using the 3009.15 A tin line and the 3010.8 copper line with spark excitation. A portion of the original alloy was analyzed similarly at the same time. The results are given in Table III below. The first two columns are the transmission readings on the tin and copper lines respectively and the third column is the ratio of the tin line intensity to that of the copper line.

TABLE III

TRANSMISSIONS		RATIO
Sn-3009.15 A	Cu-3010.8 A	Sn-3009.15 / Cu-3010.8
	On Portion of Sample Subjected to Pressure	
19.7	15.9	0.89
17.8	11.9	0.82
23.2	14.5	0.78
32.9	25.1	0.84
21.8	15.7	0.84
22.7	20.5	0.94
28.8	23.1	0.97
21.5	18.4	0.92
	Average	0.865
	On Untreated Sample-Standard	
16.0	8.7	0.75
20.6	11.2	0.73
23.4	12.7	0.72
22.0	12.2	0.74
17.0	9.5	0.76
	Average	0.74
	On Uncompressed Portion of Sample	
24.4	21.5	0.92
32.0	21.0	0.77
19.7	15.9	0.89
	Average	0.86

These results would indicate that, opposite to expectation, the element with the larger atomic volume (tin) had concentrated in the high pressure zone. This we do not believe to be the case. It is quite possible that the high ratio of the tin to the copper line intensity in this region is due, not to the higher concentration of tin, but to the different physical conditions of that portion of the specimen that has been subjected to compression.

2. The Temperature Effect

An attempt was made to reproduce the results of Darken and Oriani (6) on an Au-Cu alloy, using the bell jar type apparatus described previously. A wire of the alloy 2 inches long and 1/16-inches in diameter was used. The hot end was maintained at approximately 500° C. The temperature of the cold end was not measured, but it was presumably no higher than 100° since it was in contact with the base of the bell jar which was being cooled by tap water. The run was continued for 112 days. The pressure in the bell jar was maintained at approximately 8 microns. (Darken and Oriani used more favorable temperature conditions -- 830° C and 435° C for 32 days.)

The first method used to section the sample for x-ray analysis was to collect the dust resulting from a saw cut across the wire. However, even though a fine jeweler's saw was used, insufficient material of the proper particle size was obtained. Specimens were therefore obtained by filing, using a No. 4 cut file. In order to prevent contamination of one specimen by another, only two opposite faces of the files, which were square in cross-sections, were used. These two faces were used in preparation of adjacent specimens and loose material was removed from the file before preparing the second specimen. The filings were collected on a clean, white card and immediately transferred to

Lindemann glass capillaries having 0.2 mm outside diameter and .01 mm walls. About 0.05 to 0.07 cm length of the original diffusion sample was removed for the preparation of each x-ray specimen. Three to four capillaries could be prepared from this amount of material. In order to avoid an excessive number of specimens for x-ray analysis, a section of the original specimen approximately 0.15 cm long was removed after each filing. Thus the specimens for x-ray analysis represented an average over about 0.06 cm spaced 0.2 cm apart.

Preliminary diffraction patterns showed considerable line broadening, presumably the result of strains introduced in forming the wire, and an annealing treatment was necessary. The capillaries containing the first series of specimens prepared by the method outlined above were sealed in 10 mm diameter glass tubes under a partial atmosphere of argon. Several of these glass tubes were then sealed in a larger glass tube also containing a partial atmosphere of argon. The samples were annealed at about 350° C for 27 hours. The furnace used did not have a temperature regulator and the temperature varied ± 10° C during this annealing period. At the end of the 27-hour anneal time, the temperature was reduced to 310° C over a period of 4 hours, and this temperature was held for 16 hours. The samples were then reduced to room temperature at a rate of 18° C per hour.

After removal from the oven, the capillaries were sealed in air. The x-ray diffraction patterns were obtained with a Debye-Scherrer camera of 114.56 mm diameter. Unfiltered copper radiation was used, and the film was mounted in the Straumanis arrangement. The only lines used in calculating lattice parameters were those in the region in which the $K\alpha_1$-$K\alpha_2$ doublet was resolved.

Correction was made for film shrinkage but not for absorption by the specimen. The values of lattice parameters were plotted against $1/2\left[\cos\theta \cot\theta + \frac{\cos^2\theta}{\theta}\right]$, the extrapolation function of Nelson and Riley (12). Figure 4 is the plot for filings No. 7; this graph is typical of the results which were obtained. It is believed that the angular position of the reflections can be determined with an accuracy of ± .05° (θ). This value was used in calculating the uncertainties in the lattice parameter. It will be seen in Figure 4 that three lines occur very near 90° (θ). The assigned value of the lattice parameter is principally determined by these lines since all others have a rather large uncertainty. It will be seen that the scatter of the points from the best straight line is less than the expected uncertainty; this may be due to an overestimate of the inaccuracy in the angular position.

Table IV shows the lattice parameters calculated for the first series of specimens and the mean distance of each specimen from the cold end of the wire.

TABLE IV

Specimen	Distance from Cold End (cm)	Lattice Parameter (A)
Filings No. 7	3.96	4.0279 ± .0004
Filings No. 9	3.74	4.0275 ± .0004
Filings No. 10	3.58	4.0274 ± .0004
Filings No. 12	3.10	4.0276 ± .0004
Filings No. 14	2.73	4.0276 ± .0004

Over approximately one-fourth the sample, the lattice parameter does not vary by an amount which is outside experimental error. It was concluded, therefore, that additional investigation of this sample was not justified.

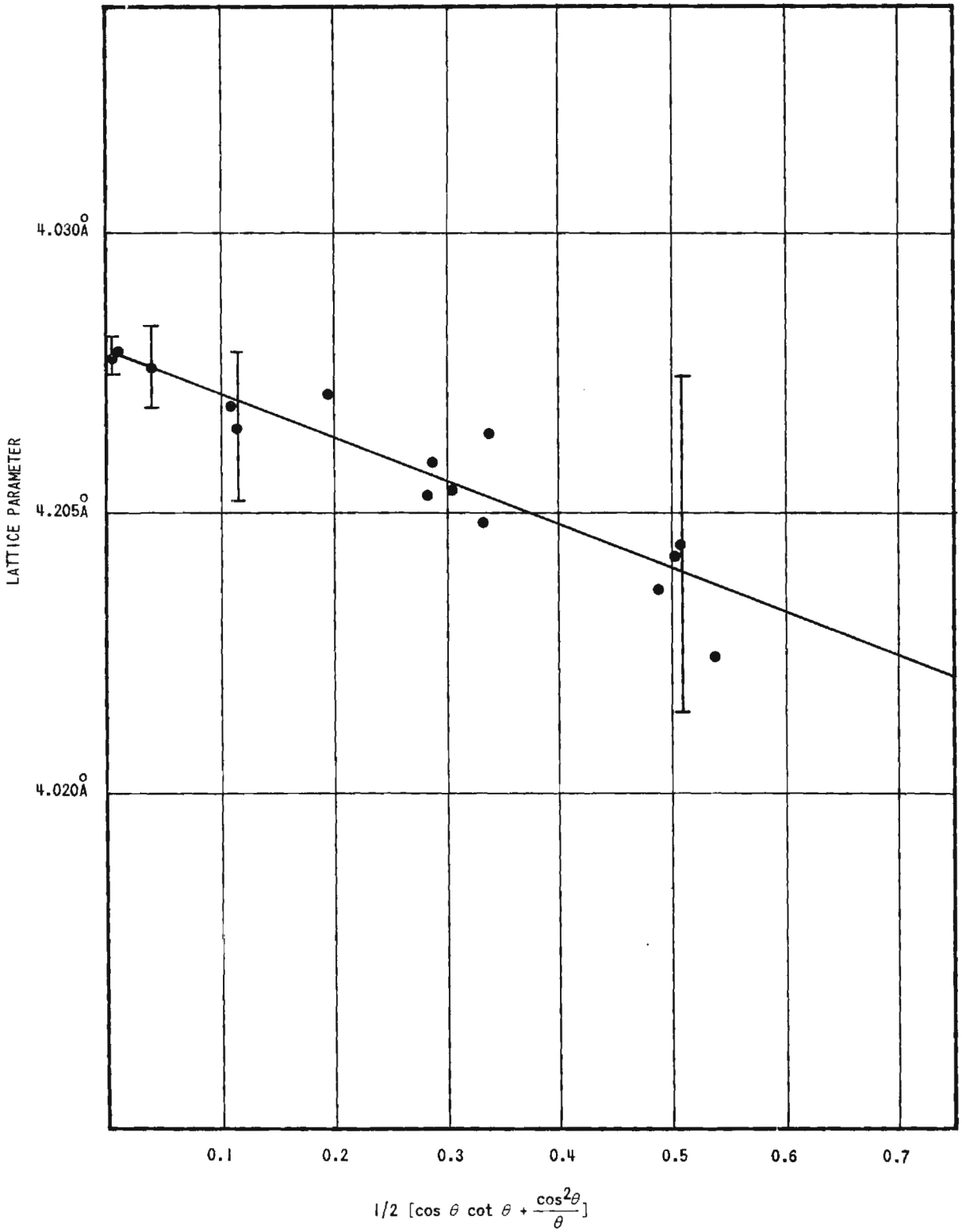

Figure 4. Gold-Copper Sample Filings.

As mentioned previously, a number of systems were investigated in an effort to demonstrate the temperature effect. Of these, the most interesting results were obtained with the three systems: Cu-0.5 per cent Cr, Pb-14 per cent Sn, and more especially, Al-21 per cent Zn. These will be discussed below. First, however, the results on the system Cu-0.4 per cent Ni alloy will be given as an example of an unsuccessful run.

a. Copper-Nickel Alloy. This was a study of the Ludwig-Soret effect, and the fact that the results were negative is perhaps not surprising since diffusion is known to be very slow in this system(7). The 1/4-inch diameter and 2-3/4-inch long rod of this alloy was held with one end at 530° C and the other at approximately 100° C for 239 days. At the end of this time, the rod was removed from the furnace and one side was ground flat. The rod was then analyzed spectrographically at nine equally spaced points from the cold to the hot end. The 3010.8 A copper and the 3002.49 A nickel lines were used with spark excitation. The results are given in Table V below.

TABLE V

TRANSMISSIONS		RATIO
Ni-3002.49	Cu-3010.8	Ni-3002.49 / Cu-3010.8
27.3	15.1	0.72
26.7	13.6	0.69
19.0	10.2	0.79
21.4	10.7	0.70
29.4	15.5	0.69
31.6	16.3	0.68
28.9	14.0	0.67
29.0	13.9	0.66
29.8	14.6	0.66

Although there is some variation in the ratio of the intensity of the nickel to the copper line, they are no greater than might be expected in the spectrographic method used.

b. Copper-Chromium Alloy. A rod of approximately 1-inch diameter of the Cu-Cr (0.08 per cent) alloy was machined as shown in Figure 5. (This figure is drawn to scale.) In order to maintain the "cold" end at a low temperature, a brass cap carrying two copper tubes was soldered on the hollow end of the specimen and tap water was circulated through this end. The specimen was placed in a furnace and maintained at such a temperature that a chromel-alumel thermocouple, which was inserted in the thermocouple wall, indicated a temperature of 750° C. The specimen was surrounded with an atmosphere of argon and the run was continued for 58 days.

At the end of the run, the specimen showed no evidence of oxidation. It was halved along its axis in order to expose a flat surface for spectrographic analysis. In this analysis, the specimen itself was used as the upper electrode and a 1/8-inch diameter graphite rod was used as the lower electrode. For the analysis, the 2979.7 A chromium and the 3010.8 A copper lines were chosen. A 5-second pre-spark and a 10-second exposure were used. The transmissions of these lines were read on a densitometer and the ratio of the intensity of the Cr to the Cu line calculated. A portion of the original alloy was used as a standard. The results are shown below.

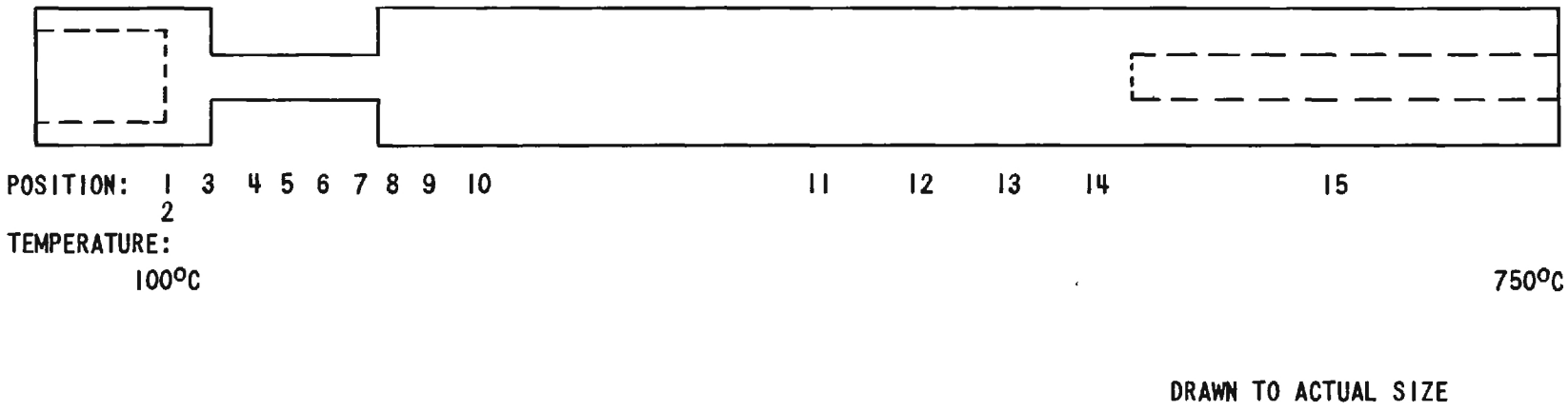

Figure 5. Copper-Chromium Alloy Sample.

RESULTS FROM FILM 1222, PLOTTED IN FIGURE 6

ROD POSITION	TRANSMISSION Cr-2979.7	TRANSMISSION Cu-3010.8	RATIO $\frac{Cr}{Cu}$
1	37.2	28.5	0.84
2	30.0	30.0	1.00
3	32.2	30.1	0.95
4	39.0	28.3	0.80
5	28.6	32.8	1.09
6	11.6	30.2	1.67
7	7.5	31.3	2.10
8	31.2	38.4	1.15
9	33.3	38.2	1.10
10	31.5	39.7	1.15
Standard	30.2	26.0	0.91
Standard	33.6	29.0	0.91
Standard	33.0	29.6	0.92

After this analysis, the specimen was polished on a cloth wheel with alundum abrasive and reanalyzed with results below.

RESULTS FROM FILM 1223, PLOTTED IN FIGURE 7

ROD POSITION	TRANSMISSION Cr-2979.7	TRANSMISSION Cu-3010.8	RATIO $\frac{Cr}{Cu}$
1	42.8	32.5	0.82
2	39.7	33.6	0.89
3	41.8	38.1	0.93
4	43.6	42.3	0.97
5	44.7	41.9	0.95
6	40.7	46.8	1.12
7	28.7	41.1	1.28
8	32.9	47.0	1.30
9	34.1	50.5	1.35
10	30.1	48.0	1.40
Standard	39.6	36.5	0.94
Standard	39.6	34.5	0.90
Standard	37.6	34.1	0.93

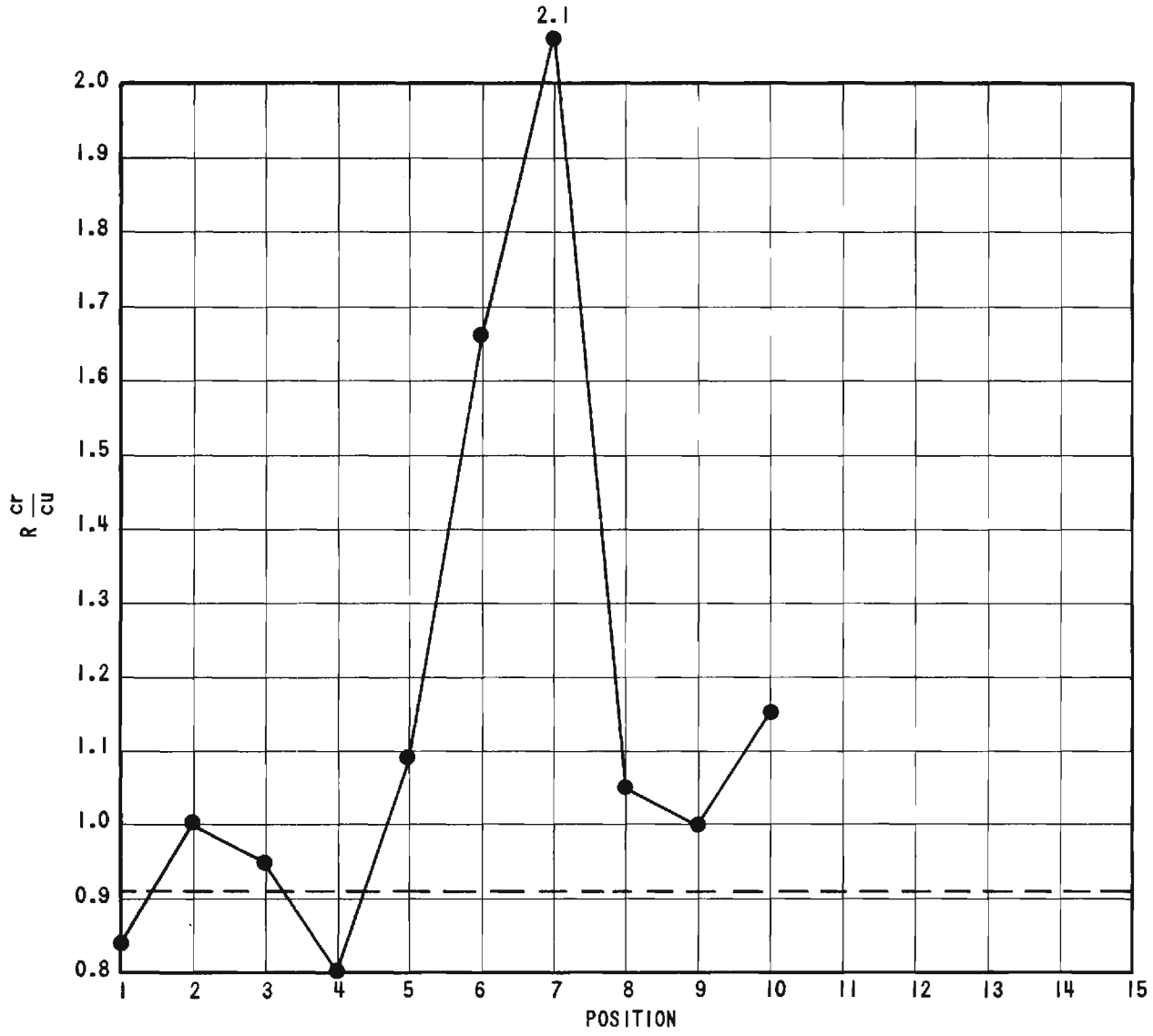

Figure 6. Ratio $\frac{Cr}{Cu}$ - Run 1.

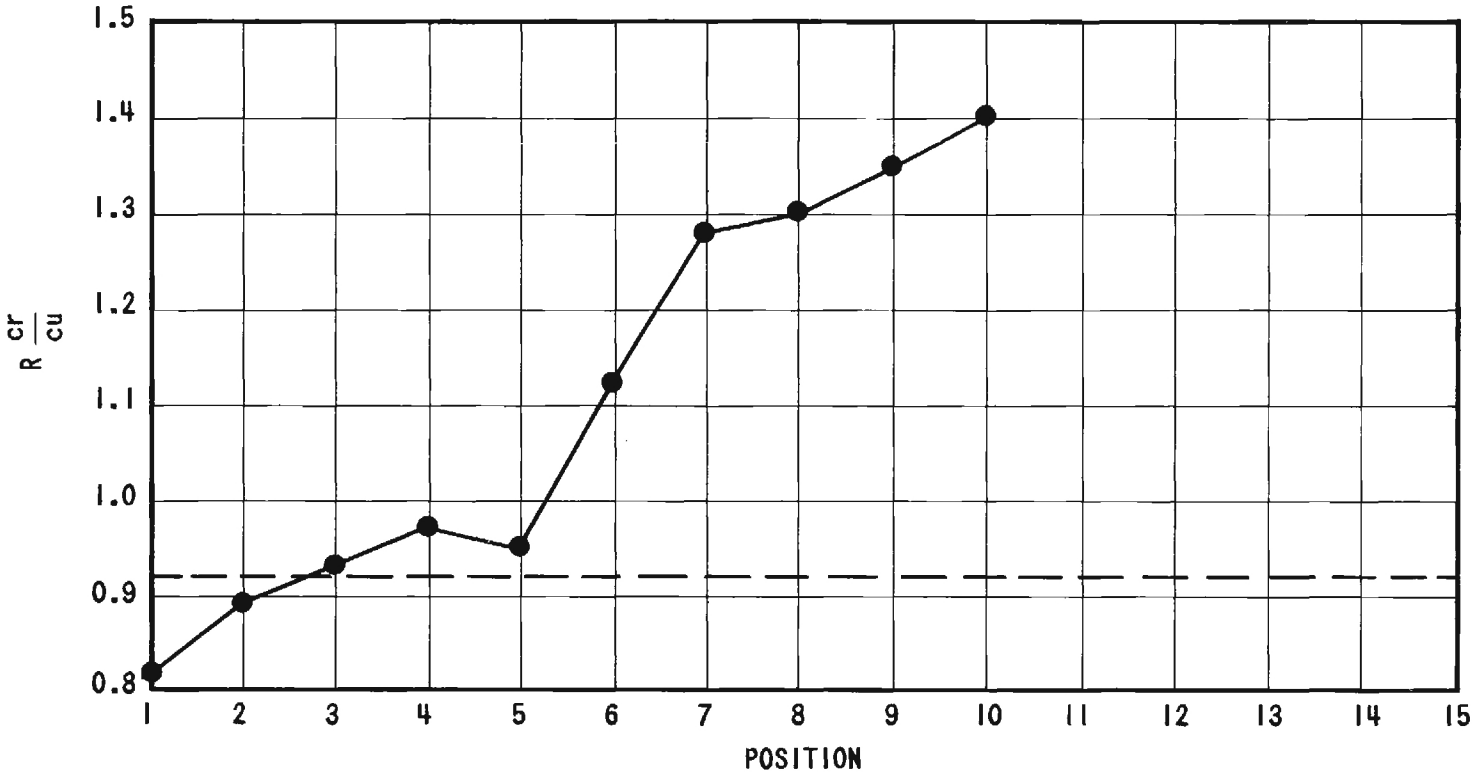

Figure 7. Ratio $\frac{Cr}{Cu}$ - Run 2.

Twice more the sample was repolished and rerun with the results below.

RESULTS FROM FILM 1224, PLOTTED IN FIGURE 8

ROD POSITION	TRANSMISSION Cr-2979.7	TRANSMISSION Cu-3010.8	RATIO $\frac{Cr}{Cu}$
1	23.4	44.0	1.54
2	20.7	38.0	1.47
3	34.2	40.0	1.10
4	35.7	42.0	1.12
5 1/2	37.2	48.5	1.22
7	25.0	49.3	1.62
8	17.2	30.0	1.40
9	25.9	55.5	1.78
10	17.5	45.4	1.85
12	19.3	36.5	1.50
12 1/2	22.0	32.0	1.25
14	13.5	38.7	1.85
15	17.6	40.0	1.67

RESULTS FROM FILM 1225, PLOTTED IN FIGURE 9

ROD POSITION	TRANSMISSION Cr-2979.7	TRANSMISSION Cu-3010.8	RATIO $\frac{Cr}{Cu}$
1	22.0	36.1	1.37
3	25.3	32.0	1.15
4	29.6	40.1	1.23
6	28.4	40.1	1.27
7	19.3	42.7	1.68
10	14.5	39.2	1.80
11	14.8	41.0	1.85
12	20.0	42.5	1.63
13 1/2	11.1	36.5	1.95
14	12.8	34.5	1.75

It will be noted that the results on films 1224 and 1225 do not agree in absolute values with one another nor with the results on films 1222 and 1223. This is not surprising since these runs were made on different days and a "curve shift" had apparently taken place. Since we had no other Cu-Cr alloys

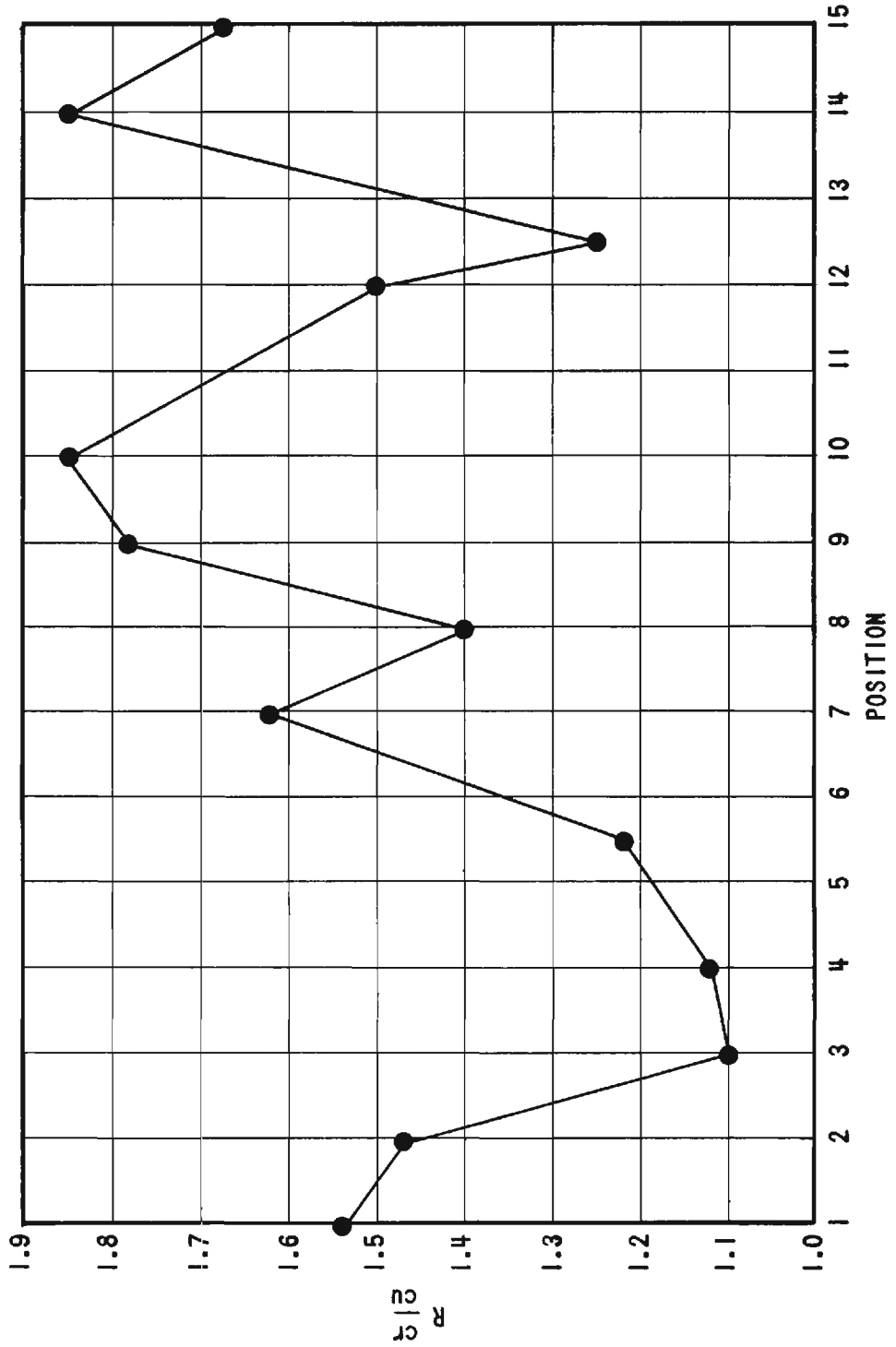

Figure 8. Ratio $\frac{Cr}{Cu}$ - Run 3.

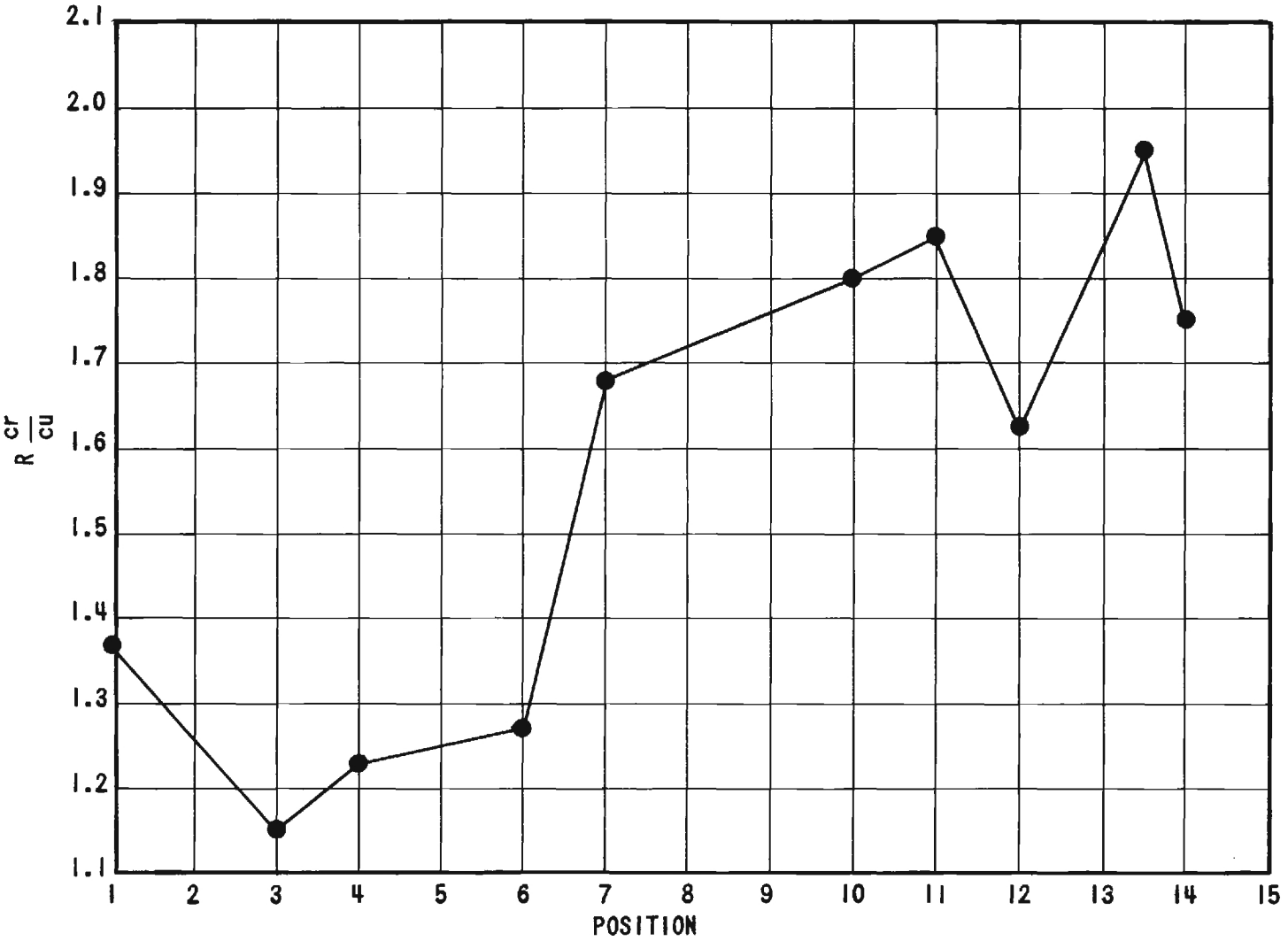

Figure 9. Ratio $\frac{Cr}{Cu}$ - Run 4.

to use as standards, the percentages of Cr corresponding to the above ratio could not be calculated. However, in a qualitative way, the ratio may be taken as a relative measure of the percentage of Cr.

It will be noted that in all four runs there was an apparent increase in concentration of Cr in the hotter portion of the specimen. That this excess Cr was extracted from the adjacent cooler portion is indicated especially well by Figures 7 and 8. It is possible that the effect varies with depth in the rod and that this explains the difference in the results shown in Figures 5 and 6 as compared with those in Figures 7 and 8.

Another run was made on this alloy in the form of a rod 1/4-inch in diameter and 2 inches long. This run lasted for 289 days with the hot end at 580° C and the cold end at approximately 100° C. The spectrographic results are given below.

RESULTS FROM FILM 1235, PLOTTED IN FIGURE 10

ROD POSITION	TRANSMISSION Cr-2979.7	TRANSMISSION Cu-3010.8	RATIO $\frac{Cr}{Cu}$
1	31.5	28.9	0.945
2	----	----	-----
3	28.7	31.8	1.07
4	----	----	-----
5	20.7	28.7	1.20
6	----	----	-----
7	22.5	26.6	1.10
8	22.6	26.7	1.10
9	----	----	-----
10	----	----	-----
Standard	34.5	25.0	0.81
Standard	36.0	27.5	0.84
Standard	33.9	27.0	0.86
Standard	30.7	20.5	0.78
Standard	31.7	23.4	0.83
Standard	33.5	24.0	0.81

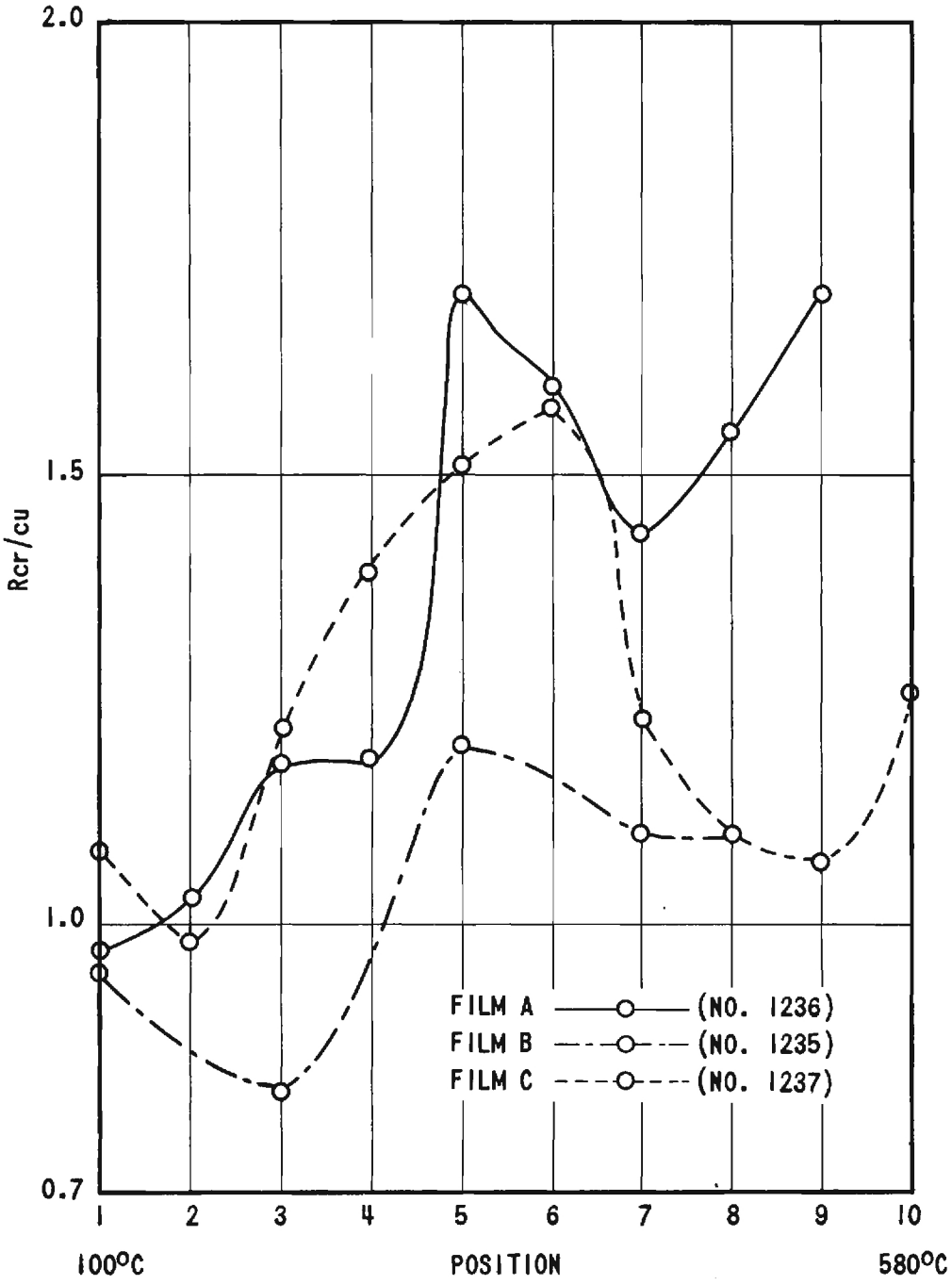

Figure 10. Cr-Cu Alloy - Time, 289 Days.

After repolishing, the rod was analyzed again.

RESULTS FROM FILM 1236, PLOTTED IN FIGURE 10

ROD POSITION	TRANSMISSION Cr-2979.7	TRANSMISSION Cu-3010.8	RATIO $\frac{Cr}{Cu}$
1	39.5	38.0	0.97
2	45.7	47.5	1.03
3	45.7	55.0	1.18
4	27.5	46.2	1.18
5	23.5	50.0	1.70
6	23.0	46.0	1.60
7	36.5	56.1	1.43
8	25.9	48.5	1.55
9	15.4	38.2	1.70
10	----	----	----
Standard	36.0	31.9	0.91
Standard	39.0	34.0	0.91
Standard	25.7	26.2	1.01
Standard	34.0	28.9	0.90

Again the rod was repolished and rerun.

RESULTS FROM FILM 1237, PLOTTED IN FIGURE 10

ROD POSITION	TRANSMISSION Cr-2979.7	TRANSMISSION Cu-3010.8	RATIO $\frac{Cr}{Cu}$
1	23.7	27.3	1.08
2	31.2	30.2	0.98
3	30.2	40.0	1.22
4	19.6	33.5	1.39
5	15.7	32.0	1.51
6	13.0	29.5	1.57
7	25.3	34.7	1.23
8	23.1	27.3	1.10
9	22.4	25.2	1.07
10	12.0	19.0	1.26

In a general way, these results agree with those obtained on the previous runs. (Plotted in Figures 6 through 9.) There definitely seemed to be an increase in the concentration of chromium in the hotter portion of the sample. These results were not pursued further, however, since the spectrographic

method was not yielding absolute results (due to lack of standards) and, by this time, the results on the Pb-Sn and the Al-Zn systems were proving more satisfactory. These will now be described.

c. Lead-Tin Alloy. A rod of this alloy 5/16-inch in diameter and 3 inches long was used. The temperature of the hot end was maintained at approximately 210° C and that of the cold end at approximately 100° C for 90 days. The rod was then cut from end to end into 45 approximately equal samples by means of a lathe. These were analyzed chemically for tin using the method of McDow, Furbee, and Clardy (9). The results are given below and plotted in Figure 11.

Sample No.	Percentage of Sn	Sample No.	Percentage of Sn
1 (Hot end)	10.23	24	14.47
2	11.00	25	14.51
3	11.58	26	14.37
4	10.97	27	14.40
5	11.81	28	14.66
6	12.43	29	-----
7	14.06	30	14.80
8	14.09	31	14.76
9	14.55	32	14.62
10	15.11	33	14.53
11	15.38	34	14.47
12	15.48	35	14.57
13	15.71	36	15.28
14	15.52	37	14.42
15	15.30	38	14.43
16	15.12	39	14.36
17	14.93	40	14.28
18	14.95	41	14.07
19	14.91	42	13.84
20	14.59	43	14.28
21	14.76	44	14.34
22	14.53	45 (Cold end)	14.52
23	14.48		

The decrease in concentration of tin below the tenth sample at the hot end was due, it is believed, to loss to the stainless steel rod into which the alloy

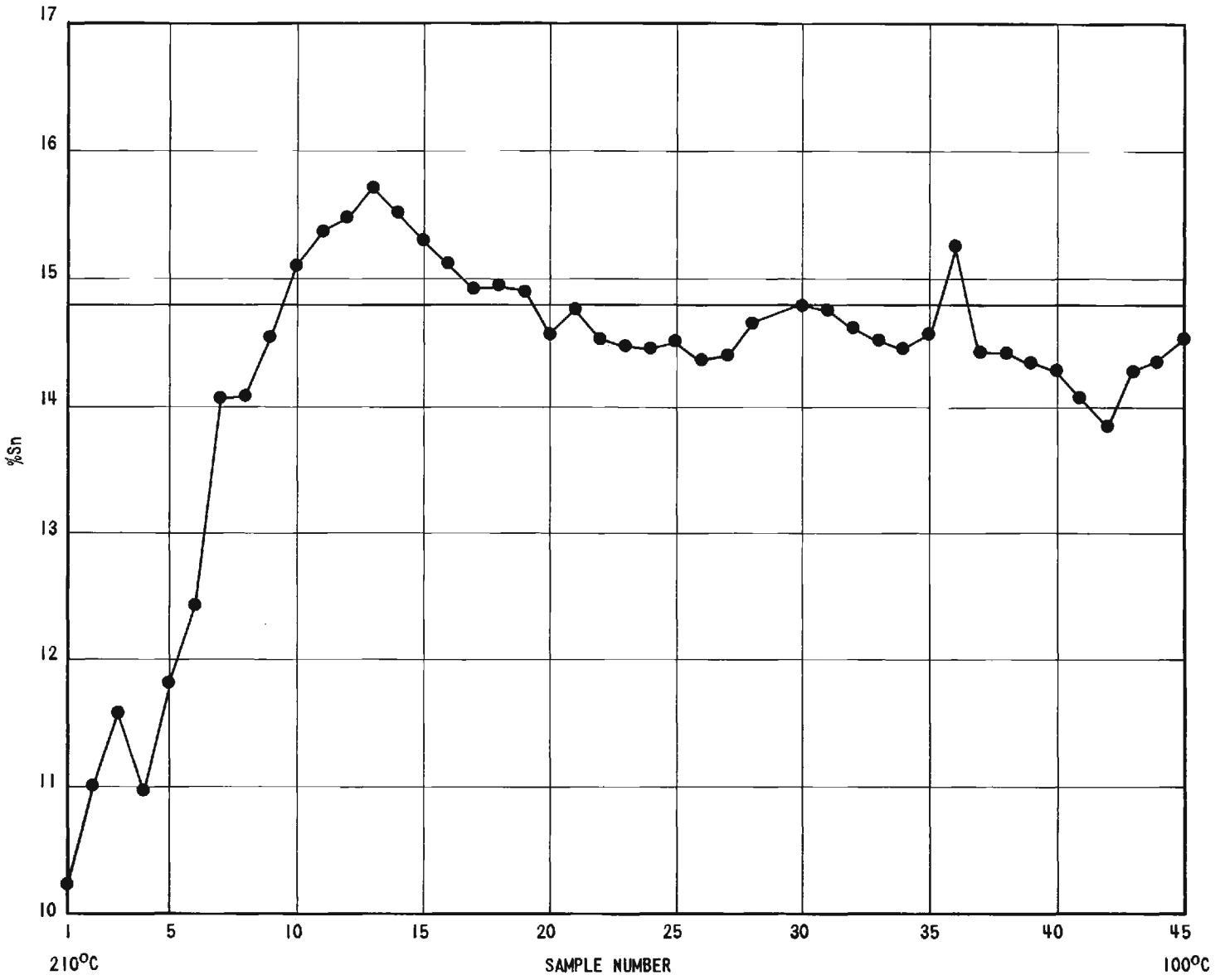

Figure 11. Sn-Pb Alloy - Time, 90 Days.

rod was screwed. (It was screwed in up through the 8th sample.)

The boundary between the saturated and the unsaturated solution regions for this concentration of tin is at approximately 170° C (10). Assuming the temperature to be 210° C at the 8th sample (the hot block of stainless steel reached this far on the Pb-Sn rod) and assuming a linear temperature distribution from here to the 45th sample, the cold end which was at about 100° C, the drop in temperature per sample would be $\frac{100° C}{37} = 3° C$. Therefore, the temperature should be 170° C at Sample No. 21. This is in good agreement with the fact that the concentration of tin begins to rise at Sample No. 19.

d. Aluminum-Zinc Alloy. Runs with this alloy were made with the following setup. A rod of the alloy 7 1/2-inches long and 3/4-inch in diameter was used. However, the middle 3 3/4-inches were machined down to 1/2-inch diameter. The purpose of this was to decrease heat conduction and thereby to increase the temperature difference along the rod. By means of a double-headed screw, the cold end was connected to a 3/4-inch diameter brass tube through which water was circulated. Thermocouples were used to measure the temperature at both the hot and cold ends of the specimen. This set up was placed in a resistance furnace as previously described.

At the end of the run, the neck of the specimen was cut on a lathe into 50 portions along its axis. Each portion was approximately 1.5 mm in length and weighed about 0.1 gram. These were analyzed using the polarographic method suggested by S. T. Payne (8). Three runs were made with results as follow.

RUN NO. 1-90 DAYS, PLOTTED IN FIGURE 12

Sample No.	Percentage of Zn	Sample No.	Percentage of Zn
1 (Hot end)	21.16	26	20.74
2	20.00	27	19.23
3	20.82	28	21.91
4	21.70	29	20.69
5	22.00	30	21.72
6	21.30	31	21.06
7	21.18	32	19.92
8	21.32	33	20.87
9	21.91	34	21.20
10	21.68	35	20.49
11	21.23	36	20.37
12	20.97	37	20.46
13	20.55	38	20.57
14	21.15	39	20.96
15	20.13	40	20.28
16	20.69	41	20.07
17	20.27	42	21.45
18	21.44	43	20.18
19	22.88	44	19.90
20	21.62	45	20.38
21	23.23	46	20.85
22	21.79	47	19.65
23	25.57	48	21.08
24	21.09	49	20.62
25	18.12	50 (Cold end)	20.85
Standard (original alloy)	20.90		

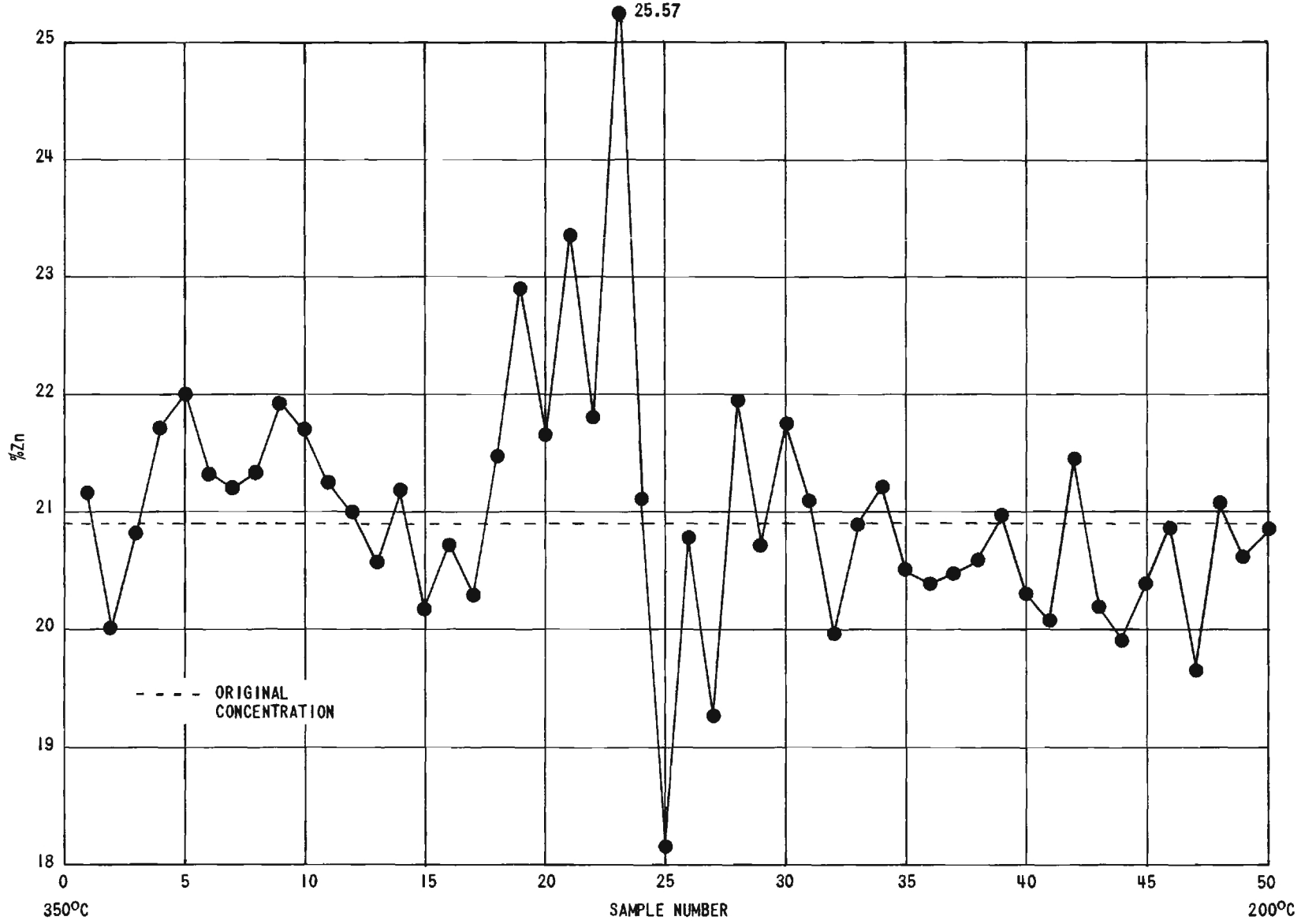

Figure 12. Zn-Al Alloy (I) - Time, 90 Days.

RUN NO. 2-137 DAYS, PLOTTED IN FIGURE 13

Sample No.	Percentage of Zn	Sample No.	Percentage of Zn
1 (Hot end)	20.20	24	20.49
2	21.05	25	21.13
3	20.87	26	19.32
4	22.39	27	20.70
5	20.63	28	20.64
6	20.41	29	20.34
7	19.20	30	20.58
8	20.78	31	19.73
9	20.29	32	20.15
10	23.82	33	20.01
11	21.92	34	20.16
12	20.39	35	20.17
13	20.29	36	20.23
14	20.33	37	20.79
15	20.58	38	21.34
16	20.38	39	20.81
17	21.44	40	20.02
18	20.46	41	19.47
19	20.24	42	21.33
20	20.67	43	20.68
21	19.46	44	20.30
22	19.96	45	20.62
23	20.92	46 (Cold end)	20.31

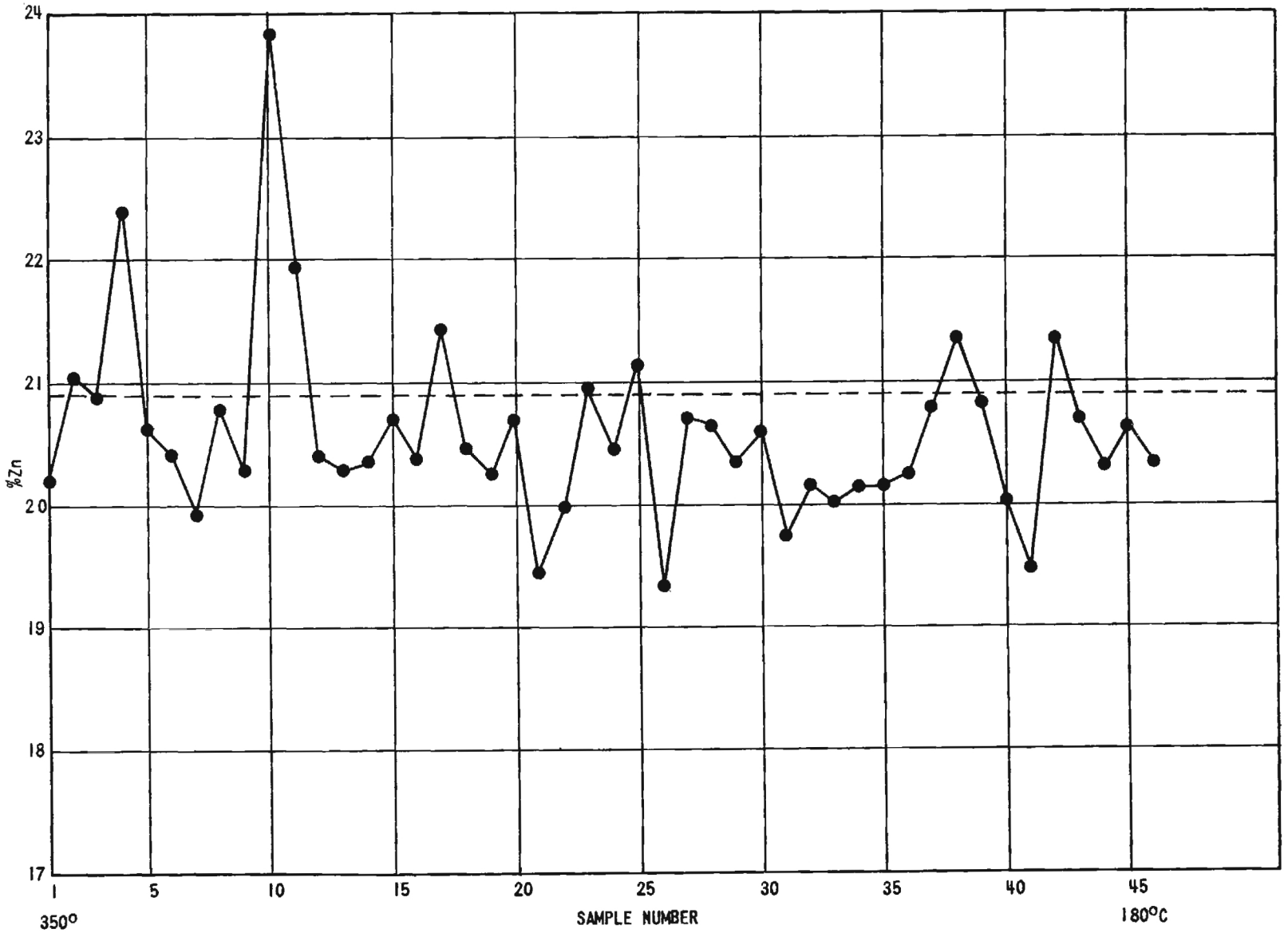

Figure 13. Zn-Al Alloy (II) - Time, 137 Days.

RUN NO. 3-158 DAYS, PLOTTED IN FIGURE 14

Sample No.	Percentage of Zn	Sample No.	Percentage of Zn
1 (Hot end)	20.41	26	20.58
2	21.10	27	20.69
3	20.75	28	20.05
4	21.33	29	20.78
5	21.20	30	20.97
6	21.28	31	20.55
7	20.79	32	19.93
8	21.23	33	17.93
9	20.39	34	20.67
10	23.87	35	20.91
11	21.63	36	18.72
12	20.25	37	20.41
13	19.62	38	20.47
14	20.37	39	19.45
15	20.10	40	21.09
16	20.96	41	20.88
17	19.74	42	21.05
18	20.59	43	21.47
19	20.57	44	20.15
20	20.67	45	20.08
21	20.73	46	20.57
22	20.21	47	21.51
23	21.94	48	20.32
24	19.97	49	20.61
25	21.53	50	20.48
		51 (Cold end)	20.35

There appears to be considerable experimental error in this method for determining zinc. In order to obtain a better idea of the size of this error,

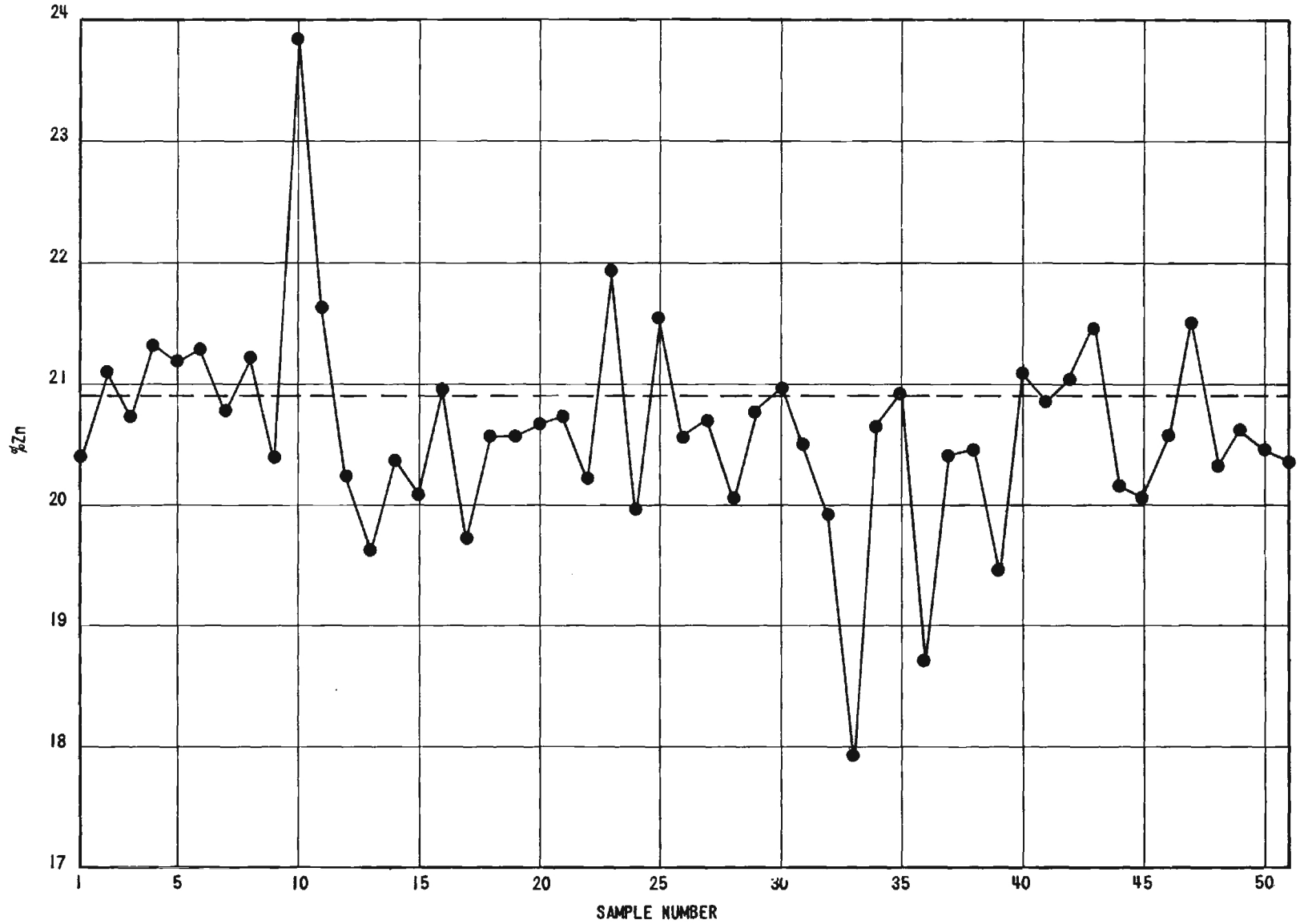

Figure 14. Zn-Al Alloy (III) - Time, 158 Days.

six samples from the turnings cut from the rod which was used for Run No. 3 (before the rod was used for the run) were analyzed in the usual way. The results which are given below confirm the fact that the error may be large.

RESULTS

Sample No.	Per Centage of Zn
1	20.80
2	20.80
3	20.76
4	20.23
5	21.65
6	19.18

In spite of this lack of good reproducibility in the method, the results of the three runs on the Al-Zn alloy definitely suggest that there is an increase in concentration of zinc towards the high temperature end of the sample. This is in agreement with our predictions and with the results on the Pb-Sn alloy, described earlier in this report. Furthermore, this increase in concentration takes place rather sharply as might be expected if it were due to crossing of the boundary between a saturated and an unsaturated solution region. This sudden increase in concentration appears at Sample No. 10 in both the second and third runs, but not until Sample No. 25 in the first run. This could well be due to a difference in the temperature distribution along the first rod as compared with the other two. That is, although the break takes place at a different sample it might be at the same temperature.

VI SUMMARY

A. The Pressure Effect

Although we failed to demonstrate this effect, we are confident that the theory is sound.

The difficulties facing a successful demonstration are:

1. The selective migration of one of the components of the alloy from the high pressure region is obscured by the relatively larger plastic flow of the alloys as a whole from this region.

2. The problem of attaining and maintaining a high pressure on a portion of the alloy is difficult to solve. A high pressure can be attained, but not maintained, by the screw-type apparatus. The dead-weight type, on the other hand, maintained the pressure but, at least in our case, it was not very high. Theoretically, of course, there is no limit to the pressure that could be attained in this way by using a large enough weight.

B. The Temperature Effect

The results on the Pb-14.8 per cent Sn alloy and on the Al-21.9 per cent Zn alloy indicate that, as predicted, when a portion of a binary alloy is maintained at a temperature where the major component is unsaturated with respect to minor component and when another portion is maintained at a temperature where the major component is saturated with the minor component, the minor com-

ponent will migrate to the region of unsaturation. Thus, a concentration gradient will develop as a result of the temperature gradient.

Respectfully submitted:

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Engineering Experiment Station

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